

FORM PTO-1300 (REV. 9-2001)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER <b>666-59079</b>	
<b>TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371</b>				U.S. APPLICATION NO. (If known, see 37 CFR 1.5 <div style="font-size: 1.5em; font-weight: bold; text-align: center;">10/019022</div>	
INTERNATIONAL APPLICATION NO. <b>PCT/EP00/05980</b>		INTERNATIONAL FILING DATE <b>27 June 2000</b>		PRIORITY DATE CLAIMED <b>29 June 1999</b>	
TITLE OF INVENTION <div style="text-align: center; font-weight: bold;">PROCESS FOR THE PRODUCTION OF ALUMINIUM HYDROXIDE</div>					
APPLICANT(S) FOR DO/EO/US <b>BILANDZIC, Genoveva; BROWN, Neil, PUTZ, Norbert</b>					
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:					
<ol style="list-style-type: none"> <li>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li>3. <input type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.</li> <li>4. <input checked="" type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31).</li> <li>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))           <ol style="list-style-type: none"> <li>a. <input checked="" type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau).</li> <li>b. <input checked="" type="checkbox"/> has been communicated by the International Bureau.</li> <li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</li> </ol> </li> <li>6. <input type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).           <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> is attached hereto.</li> <li>b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4).</li> </ol> </li> <li>7. <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))           <ol style="list-style-type: none"> <li>a. <input checked="" type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau).</li> <li>b. <input checked="" type="checkbox"/> have been communicated by the International Bureau.</li> <li>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li>d. <input type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li>8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).</li> <li>9. <input type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</li> <li>10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</li> </ol>					
<b>Items 11 to 20 below concern document(s) or information included:</b>					
<ol style="list-style-type: none"> <li>11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</li> <li>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</li> <li>13. <input checked="" type="checkbox"/> A FIRST preliminary amendment to be entered prior to claims fee calculation.</li> <li>14. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment.</li> <li>15. <input type="checkbox"/> A substitute specification.</li> <li>16. <input type="checkbox"/> A change of power of attorney and/or address letter.</li> <li>17. <input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.</li> <li>18. <input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4).</li> <li>19. <input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).</li> <li>20. <input checked="" type="checkbox"/> Other items or information: <b>International Search Report International Preliminary Examination Report Form PCT/IB/306; Form PCT/IB/332; Form PCT/IB/308</b></li> </ol>					

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re PATENT Application of  
Bilandzic, et. al.

Group Art Unit: Unknown

U.S. National Phase of PCT/EP00/05980

Serial No.: Unknown

Examiner: Unknown

Filed: December 26, 2001

For: PROCESS FOR THE PRODUCTION OF ALUMINUM HYDROXIDE

\* \* \* \* \*

December 26, 2001

**PRELIMINARY AMENDMENT TO BE CONSIDERED PART OF ORIGINAL FILING  
AND ENTERED PRIOR TO CLAIMS FEE CALCULATION**

Hon. Asst. Commissioner of Patents  
and Trademarks  
Washington, D.C. 20231

Sir:

Please preliminarily amend the subject application prior to calculation of the claims fees as follows:

IN THE CLAIMS:

Please amend claims 1-5 and 7-9 as follows:

1. (Amended) A process for the preparation of gibbsite type aluminum hydroxides comprising:  
seeding a liquor obtained from a Bayer process with bayerite crystals having a grain diameter in the 50% range ( $d_{50}$ ) of 1.2  $\mu\text{m}$  to 2.2  $\mu\text{m}$ .
2. (Amended) A process according to claim 1, wherein the molar ratio of  $\text{Na}_2\text{O}$  to  $\text{Al}_2\text{O}_3$  in the Bayer liquor is from 1.4:1 to 1.55:1.
3. (Amended) A process according to claim 1, wherein the bayerite crystals have a grain diameter in the 10% range ( $d_{10}$ ) of 0.5  $\mu\text{m}$  to 0.9  $\mu\text{m}$  and in the 90% range ( $d_{90}$ ) of 3.5  $\mu\text{m}$  to 5.5  $\mu\text{m}$ .
4. (Amended) A process according to claim 1, wherein the bayerite crystals are added in an amount of 1 g/l to 3 g/l at a liquor temperature of 50°C to 70°C.

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5. (Amended) A process according to claim 1, wherein after seeding the process mixture is stirred under precipitation until the molar ratio of  $\text{Na}_2\text{O}$  to  $\text{Al}_2\text{O}_3$  is in the range of 2.2:1 to 3.5:1.
7. (Amended) A mixture of gibbsite type aluminum hydroxides, wherein the mixture has a grain diameter in the 50% range of ( $d_{50}$ ) 4 to 20  $\mu\text{m}$  and comprises a gibbsite type aluminum hydroxide according to claim 6.
8. (Amended) A synthetic resin comprising a gibbsite type aluminum hydroxide according to claim 6.
9. (Amended) The use of a gibbsite type aluminum hydroxide according to claim 6 as a fire retardant additive in synthetic resins.

#### EXPLANATION OF AMENDMENT:

The claims have been amended as shown by [deletions] and insertions.

1. (Amended) A process for the preparation of gibbsite type aluminum hydroxides[, wherein] comprising  
seeding a liquor obtained from [the] a Bayer process [is seeded] with bayerite crystals having a grain diameter in the 50% range ( $d_{50}$ ) of 1.2  $\mu\text{m}$  to 2.2  $\mu\text{m}$ .
2. (Amended) [The] A process [of] according to claim 1, wherein the molar ratio of  $\text{Na}_2\text{O}$  to  $\text{Al}_2\text{O}_3$  in the Bayer liquor is from 1.4:1 to 1.55:1.
3. (Amended) A process according to claim 1 [any one of claims 1 to 2], wherein the bayerite crystals have a grain diameter in the 10% range ( $d_{10}$ ) of 0.5  $\mu\text{m}$  to 0.9  $\mu\text{m}$  and in the 90% range ( $d_{90}$ ) of 3.5  $\mu\text{m}$  to 5.5  $\mu\text{m}$ .
4. (Amended) A process according to claim 1 [any one of claims 1 to 3], wherein the bayerite crystals are added in an amount of 1 g/l to 3 g/l at a liquor temperature of 50°C to 70°C.
5. (Amended) A process according to claim 1 [any one of claims 1 to 4], wherein after seeding the process mixture is stirred under precipitation until the molar ratio of  $\text{Na}_2\text{O}$  to  $\text{Al}_2\text{O}_3$  is in the range of 2.2:1 to 3.5:1.
7. (Amended) A mixture of gibbsite type aluminum hydroxides, [characterised in that it] wherein the mixture has a grain diameter in the 50% range of ( $d_{50}$ ) 4 to 20  $\mu\text{m}$  and [contains] comprises a gibbsite type aluminum hydroxide according to claim 6 [or a gibbsite type aluminum hydroxide obtained by a

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process according to any one of claims 1 to 5].

8. (Amended) A synthetic resin [containing] comprising a gibbsite type aluminum hydroxide according to claim 6 [or obtained by a process according to any one of claims 1 to 5].
9. (Amended) The use of a gibbsite type aluminum hydroxide according to claim 6 [or obtained by a process according to any one of claims 1 to 5] as a fire retardant additive in synthetic resins.

#### REMARKS

Consideration and allowance of the subject application are respectfully requested.

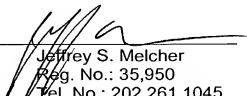
Claims 1-9 are pending in the application.

Claims 1-5 and 7-9 have been amended to correct minor informalities. No claims have been amended to overcome prior art. No new matter has been added.

Entry of this amendment and a favorable action on the merits are respectfully requested.

Respectfully submitted,  
Manelli Denison & Selter, PLLC

By

  
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Process for the production of aluminium hydroxide

The invention relates to a process for the production of aluminium hydroxide.

- 5 Aluminium hydroxide is widely used as fire retardant filler in glass reinforced plastic composite materials, the fire safety of which is an issue of increasing importance.

In order to meet the increasingly stringent fire safety requirements, it is important to be able to raise the filling level of aluminium hydroxide to ever higher values without  
10 compromising the physical properties of the resulting composite material.

It is well-known in the art that increased filling levels can be achieved by mixing aluminium hydroxides having different particle size distributions in order to obtain a more densely packed dispersion in synthetic resin (Rai et al. *Inorganic Flame Retardants—Alone*  
15 *and in combinations*, Presentation at *Flame Retardants 98*, February 1998, London). In general, the wider the resulting particle size distribution (psd), the lower the viscosity and, hence, the better the workability of the dispersion. However, the limitations here are that coarser particles of aluminium hydroxide will result in poorer surface quality of the finished part and degradation of mechanical properties.

20

The challenge to the industry is to achieve high filling levels at finer particle sizes. In addition, it is particle size and particle shape which combine to give the best possible packing so that only by exploiting the latter optimum results can be obtained.

- 25 The problem here is that the finest particles themselves are limited in terms of complexity of particle shape adopted during crystallisation. The alternative is grinding to very fine particle sizes, but this also has limited use because the crystallinity of the material is decreased and high energy surfaces are formed which create additional problems of high moisture content and interference with the chemistry of synthetic resins and their curing  
30 mechanisms.

Three forms of aluminium hydroxide are known: gibbsite ( $\alpha$ -aluminium hydroxide), bayerite ( $\beta$ -aluminium hydroxide) and nordstrandite (K. Wefers and C. Misra *Oxides and Hydroxides of Aluminium*; Alcoa Technical Paper No. 19, Revised: 1987). Of these, gibbsite is the best known and occurs in nature as the principal constituent of bauxite.

- 5 Synthetic gibbsite is an intermediate of the Bayer process for the production of alumina from bauxite. It is prepared by seeding supersaturated sodium aluminate solution with previously formed gibbsite crystals.

- 10 Bayerite does not occur in nature but it can be made in various ways, e. g., by treating sodium aluminate solution with carbon dioxide at about 20 °C. However, a key property of bayerite is its transformation to the more thermodynamically stable phase, gibbsite, on prolonged holding under such strongly alkaline conditions (K. Wefers and C. Misra *loc. cit.*).

- 15 Nordstrandite rarely occurs naturally, but can be obtained synthetically from gelatinous hydroxide by ageing in the presence of a chelating agent such as ethylene diamine, ethylene glycol or EDTA.

- 20 Of the three forms of aluminium hydroxide, gibbsite is technically the most important. It is the preferred form used by the plastics industry to impart fire retardant properties to composite materials.

- 25 The crystallisation of aluminium hydroxide by the Bayer process is well-known in the art. This aluminium hydroxide however is destined for calcination to alumina and subsequent smelting to aluminium metal. Its coarse particles are not suitable for use as a flame retardant in plastics.

- 30 At the other end of the particle size spectrum are the fine crystals which are generally produced in a side stream process in which the crystallisation conditions are altered to create new crystals by a secondary nucleation mechanism, i. e., the seed crystals are very fine and specially prepared to be active in generating new nuclei.

The large gap in the middle of the particle size spectrum, comprising a particle size of about 4 to 20  $\mu\text{m}$ , is normally covered via size reduction of the very large particles obtained in the crystallisation step of the Bayer process. Thereafter, mixtures of differing psd can be produced by mixing to achieve improved packing characteristics which lead to lower viscosities in synthetic resins, and hence higher obtainable filling levels. However, grinding to an average particle size of less than 5  $\mu\text{m}$  increasingly creates only crystalline debris with distorted surfaces.

A difficulty limiting the use of very fine crystals is their pronounced tendency to aggregate and agglomerate during crystallisation. These effects are deleterious to good rheology in synthetic resins since they can increase viscosity levels dramatically. When mixed with coarser crystals to broaden the psd, they cannot be incorporated at levels which can exploit their fireproofing properties.

The object of the present invention therefore is to provide a process for the production of a fine crystalline aluminium hydroxide which does not give rise to the drawbacks outlined above and known from prior art.

This object could be achieved by the process of claim 1, the gibbsite type aluminium hydroxide obtained by said process according to claim 7, and the synthetic resin filled with the respective aluminium hydroxide according to claim 10.

Surprisingly, it has been found that during particle size enlargement via crystal growth gibbsite crystals nucleated by bayerite seed retain their characteristic platy pseudo-hexagonal crystal habit much longer than do fine crystals nucleated by gibbsite (highly active, finely ground seed).

Also standard crystallisation measures taken to further increase the size of the crystals, e. g., raising the temperature and/or reducing the seed charge increase the size of the crystals with surprisingly little aggregation and/or agglomeration, which is the normal occurrence with gibbsite crystals.



The process of the present invention is characterised in that a liquor obtained from the Bayer process is seeded with bayerite crystals and subsequently stirred under precipitation of the gibbsite type aluminium hydroxide. Thereafter the solid obtained in suspension is filtered off.

5

The liquor from the Bayer process can be defined by a molar ratio of  $\text{Na}_2\text{O}$  to  $\text{Al}_2\text{O}_3$  in the range of 1.4:1 to 1.55:1, preferably from 1.45:1 to 1.50:1.

10 The bayerite crystals used as seed expediently have a grain diameter in the 50% range ( $d_{50}$ ) of 1.2  $\mu\text{m}$  to 2.2  $\mu\text{m}$ , preferably of 1.5  $\mu\text{m}$  to 1.8  $\mu\text{m}$ . They can further be characterised by a grain diameter in the 10% range ( $d_{10}$ ) of 0.5  $\mu\text{m}$  to 0.9  $\mu\text{m}$ , preferably 0.6  $\mu\text{m}$  to 0.8  $\mu\text{m}$ , and a grain diameter in the 90% range ( $d_{90}$ ) of 3.5  $\mu\text{m}$  to 5.5  $\mu\text{m}$ , preferably 4.0  $\mu\text{m}$  to 4.5  $\mu\text{m}$ .

15 The bayerite crystals used for the process according to the invention may be obtained by seeding a pregnant (supersaturated) Bayer liquor having a molar ratio of  $\text{Na}_2\text{O}$  to  $\text{Al}_2\text{O}_3$  as given above and having been cooled to a temperature of 20 °C to 25 °C with 2 g/l to 3 g/l of bayerite crystals previously obtained.

20 The resulting suspension as a rule will be stirred for a period of 15 to 25 hours, but at least until the solid content has risen to 60 g/l to 120 g/l.

The crystals in this suspension have an average particle size of 5 to 15  $\mu\text{m}$  and a specific surface area (according to BET) of 5 to 10  $\text{m}^2/\text{g}$ .

25 After filtering, washing the crystals with water and resuspending them in water, the suspension undergoes a size reduction treatment to lower the average crystal size to less than 1  $\mu\text{m}$  and to raise the specific surface area (BET) to more than 20  $\text{m}^2/\text{g}$ .

The size reduction treatment may be brought about in mills customarily employed for this kind of milling, e. g., bead mills.

The crystals obtained are characterised as described above.

Seeding as a rule is effected by adding the bayerite crystals obtained to the Bayer liquor at a temperature of 50 °C to 70 °C and in an amount (relating to the Bayer liquor) of 1 g/l to 3 g/l.

During crystallisation the bayerite seed is fully transformed to gibbsite.

5

Precipitation usually takes place under stirring and will be continued until the molar ratio of  $\text{Na}_2\text{O}$  to  $\text{Al}_2\text{O}_3$  is in the range of 2.2:1 to 3.5:1, preferably from 2.5:1 to 3.0:1.

The precipitated gibbsite type aluminium hydroxide can easily be collected by filtration.

10

The gibbsite type crystalline aluminium hydroxide prepared by the process of the present invention typically has the following characteristics:

- A grain diameter in the 50% range ( $d_{50}$ ) of 1.5  $\mu\text{m}$  to 4.5  $\mu\text{m}$ , preferably 1.8  $\mu\text{m}$  to 4.0  $\mu\text{m}$ , in the 10% range ( $d_{10}$ ) of 0.4  $\mu\text{m}$  to 2.0  $\mu\text{m}$ , preferably 0.5  $\mu\text{m}$  to 1.8  $\mu\text{m}$ , and
- 15 in the 90% range ( $d_{90}$ ) of 3.0  $\mu\text{m}$  to 9.5  $\mu\text{m}$ , preferably 3.5  $\mu\text{m}$  to 9.0  $\mu\text{m}$ .
- A specific surface area (BET) in the range of 1.0  $\text{m}^2/\text{g}$  to 4.0  $\text{m}^2/\text{g}$ , preferably 2.0  $\text{m}^2/\text{g}$  to 3.0  $\text{m}^2/\text{g}$ .
- An aspect ratio (i. e., the ratio length:thickness) of a crystal of about 4:1 to 6:1.

20

The crystalline gibbsite type aluminium hydroxide obtained by the process of the present invention can — either alone or as part of a mixture — be filled into synthetic resins, preferably into thermosetting plastics, such as unsaturated polyester or epoxy resins. Extraordinarily high filling levels of 150 phr (parts per hundred parts of resin) to 200 phr can be achieved.

25

In order to achieve high filling levels at finer particle sizes usually mixtures of aluminium hydroxides having different particle size distributions are applied. The gibbsite type aluminium hydroxide crystals obtained by the process of the invention are ideally suited to be mixed with materials obtained by grinding coarse aluminium hydroxide produced by the

30

Bayer process. On size reduction (grinding), coarse aluminium hydroxides increasingly

adopt a plate like form due to cleavage parallel to the basal planes of the blocky hexagonal prisms. Preferably the mixtures have a grain diameter in the 50% range ( $d_{50}$ ) of 4 to 20  $\mu\text{m}$ .

The following non-limiting examples illustrate the invention.

- 5 All MARTINAL<sup>®</sup> types of aluminium hydroxide are commercially available from Alu-suisse Martinswerk GmbH, Bergheim/Erft, Germany. The Synolite<sup>®</sup> resins are products of DSM-BASF Structural Resins. All viscosity measurements were performed at 23 °C in a Brookfield HBT viscosimeter with the spindle and the rotational speed (given in  $\text{min}^{-1}$ ,  $1 \text{ min}^{-1} = 1 \text{ revolution per minute}$ ) specified in the column "viscosimeter settings" of the
- 10 corresponding table.

### Example 1

- 15 A sodium aluminate solution having a temperature of 60 °C and containing 140 g/l of  $\text{Na}_2\text{O}$ , 150 g/l of  $\text{Al}_2\text{O}_3$  and 160 g/l of total soda was seeded with 2 g/l of bayerite crystals having the following characteristics:

$$d_{50} = 1.6 \mu\text{m}$$

$$d_{10} = 0.75 \mu\text{m}$$

20  $d_{90} = 4.25 \mu\text{m}$

- The bayerite crystals had previously been obtained by seeding a sodium aluminate solution having a temperature of 20 to 22 °C and containing 140 g/l of  $\text{Na}_2\text{O}$ , 155 g/l of  $\text{Al}_2\text{O}_3$  and 160 g/l of total soda (molar ratio  $\text{Na}_2\text{O}$  to  $\text{Al}_2\text{O}_3 = 1.48$ ) with 2 to 3 g/l of bayerite crystals. The suspension was stirred for 20 hours until the solids content has risen to 100 g/l. The
- 25 suspended bayerite crystals had a grain diameter in the 50% range ( $d_{50}$ ) of 1.6  $\mu\text{m}$  and a BET surface of 3.4  $\text{m}^2/\text{g}$ . After filtering the suspension, washing the crystals with water and resuspending the crystals in water (concentration ca. 100 g/l), size reduction in a bead mill was accomplished.

- 30 Mill parameter:

Mill type: Backofen

Capacity: 200 l  
 Beads: 1.5 mm diameter  
 Total Charge: 80%  
 Throughput: 2000-3000 l/h

5

The crystals thus obtained were filtered off. Their characteristics were as hereinabove described.

After a retention time of 48 hours at 60 °C, 100 g/l of gibbsite type aluminium hydroxide was obtained and the bayerite seed was fully transformed to gibbsite during the

10 crystallisation period.

#### Examples 2-6, Comparative Examples 1-2

15 The procedure of Example 1 was repeated with various seed charges and crystallization temperatures.

The comparative examples were performed following the same procedure as described above, with the exception that gibbsite was used as seed material.

20 Table 1 shows the influence of these parameters on the grain diameter in the 50% range ( $d_{50}$ ) of the gibbsite type aluminium hydroxide obtained and the largest particle size present ("top cut") in Examples 1-6 and Comparative Examples 1-2.

Table 1

Example No.	Seed Charge [g/l]	Cryst. Temp. [°C]	$d_{50}$ [μm]	Top Cut [μm]
1	1	60	2.8	7
2	2	60	1.8	6
3	3	60	1.6	6
4	1	65	3.6	8
5	1	70	4.1	9
6	2	70	3.5	8
Comp. 1	8	60	1.4	7
Comp. 2	6	60	2.1	9

**Examples 7–8, Comparative Examples 3–4**

The effect of the different product forms on rheology was tested in synthetic resin. 150 parts per hundred (phr) of the product of Examples 2 and 6 and Comparative Examples 1 and 2 were introduced in the unsaturated polyester resin Synolite® 002-N-2. The results are compiled in Table 2.

**Table 2**

Example No.	Product source	Viscosity [mPa·s]	Viscosimeter settings
7	Ex. 3	60	spindle 3 @ 5 min <sup>-1</sup>
8	Ex. 6	70	spindle 4 @ 10 min <sup>-1</sup>
Comp. 3	Comp. Ex. 1	750	spindle 7 @ 10 min <sup>-1</sup>
Comp. 4	Comp. Ex. 2	1500	spindle 7 @ 10 min <sup>-1</sup>

10

**Example 9, Comparative Example 5**

Following the procedure of Examples 7–8, rheology in synthetic resin was tested with a mixture of gibbsite type aluminium hydroxide obtained according to Example 2 and coarse size reduced aluminium hydroxide crystals obtained from the Bayer process. Comparison was made against a mixture of the fine gibbsite seeded aluminium hydroxide obtained according to Comparative Example 1 and coarse size reduced aluminium hydroxide crystals obtained from the Bayer process.

175 parts per hundred (phr) of a 1:1 mixture ( $d_{50} = 4 \mu\text{m}$ ) of the product of Example 2 or Comparative Example 1 and MARTINAL® ON-310 ( $d_{50} = 10 \mu\text{m}$ ) was introduced in the unsaturated polyester resin Synolite® 002-N-2. The results are shown in Table 3.

Table 3

Example No.	Product source	Viscosity [mPa·s]	Viscosimeter settings
9	Ex. 2	65	spindle 5 @ 20 min <sup>-1</sup>
Comp. 5	Comp. Ex. 1	220	spindle 6 @ 10 min <sup>-1</sup>

**Example 10, Comparative Example 6**

175 parts per hundred (phr) of a 1:2 mixture ( $d_{50} = 14 \mu\text{m}$ ) of the product of Example 6 and MARTINAL® ON-320 ( $d_{50} = 20 \mu\text{m}$ ) was introduced in the unsaturated polyester resin Synolite 002-N-2. In Comparative Example 6, the product of Example 6 was replaced with the product of Comparative Example 1

10 The results are shown in Table 4.

Table 4

Example No.	Product source	Viscosity [mPa·s]	Viscosimeter settings
10	Ex. 6	28	spindle 5 @ 50 min <sup>-1</sup>
Comp. 6	Comp. Ex. 1	42	spindle 3 @ 5 min <sup>-1</sup>

**Example 11, Comparative Example 7**

175 parts per hundred (phr) of a 1:1:1 mixture ( $d_{50} = 18 \mu\text{m}$ ) of (i) the product of Example 1, (ii) ground grade MARTINAL® ON-320 ( $d_{50} = 20 \mu\text{m}$ ) and (iii) unground grade ( $d_{50} = 55 \mu\text{m}$ ) MARTINAL® ON was introduced into the Synolite® 002-N-2 unsaturated polyester resin.

The viscosity of this mix was compared with that of another mix in which the product of Example 1 was replaced with the product of Comparative Example 2. The results are given in Table 5.

Table 5

Example No.	Product source	Viscosity [mPa·s]	Viscosimeter settings
11	Ex. 1	20	spindle 5 @ 50 min <sup>-1</sup>
Comp. 7	Comp. Ex. 2	29	spindle 5 @ 50 min <sup>-1</sup>

## Example 12, Comparative Example 8

5

125 parts per hundred (phr) of a 1:3 mixture ( $d_{50} = 16\mu\text{m}$ ) of the product of Example 4 ( $d_{50} = 3.6\mu\text{m}$ ) and the milled grade ( $d_{50} = 20\mu\text{m}$ ) MARTINAL<sup>®</sup> ON-320 was introduced into the epoxy resin Araldite<sup>®</sup> GY 260 (Ciba Polymers). The viscosity of this mix was compared with another in which the product of Example 4 was replaced with the product of

10 Comparative Example 2. The results are shown in Table 6.

Table 6

Example No.	Product source	Viscosity [mPa·s]	Viscosimeter settings
12	Ex. 4	82	spindle 5 @ 10 min <sup>-1</sup>
Comp. 8	Comp. Ex. 2	140	spindle 5 @ 10 min <sup>-1</sup>

23-07-2001

10/019022  
JC13 Rec'd PCT/PTO 26 DEC 2001

## Claims

1. A process for the preparation of gibbsite type aluminium hydroxides, wherein a liquor obtained from the Bayer process is seeded with bayerite crystals having a grain diameter in the 50% range ( $d_{50}$ ) of 1.2  $\mu\text{m}$  to 2.2  $\mu\text{m}$ .
2. The process of claim 1, wherein the molar ratio of  $\text{Na}_2\text{O}$  to  $\text{Al}_2\text{O}_3$  in the Bayer liquor is from 1.4:1 to 1.55:1.
3. A process according to any one of claims 1 to 2, wherein the bayerite crystals have a grain diameter in the 10% range ( $d_{10}$ ) of 0.5  $\mu\text{m}$  to 0.9  $\mu\text{m}$  and in the 90% range ( $d_{90}$ ) of 3.5  $\mu\text{m}$  to 5.5  $\mu\text{m}$ .
4. A process according to any one of claims 1 to 3, wherein the bayerite crystals are added in an amount of 1 g/l to 3 g/l at a liquor temperature of 50 °C to 70 °C.
5. A process according to any one of claims 1 to 4, wherein after seeding the process mixture is stirred under precipitation until the molar ratio of  $\text{Na}_2\text{O}$  to  $\text{Al}_2\text{O}_3$  is in the range of 2.2:1 to 3.5:1.
6. A gibbsite type aluminium hydroxide having a grain diameter in the 50% range ( $d_{50}$ ) of 1.5  $\mu\text{m}$  to 4.5  $\mu\text{m}$ , in the 10% range ( $d_{10}$ ) of 0.4  $\mu\text{m}$  to 2.0  $\mu\text{m}$  and in the 90% range ( $d_{90}$ ) of 3.0  $\mu\text{m}$  to 9.5  $\mu\text{m}$ , a surface area (according to BET) from 1.0  $\text{m}^2/\text{g}$  to 4.0  $\text{m}^2/\text{g}$  and an aspect ratio from 4:1 to 6:1.
7. A mixture of gibbsite type aluminium hydroxides, characterised in that it has a grain diameter in the 50% range of ( $d_{50}$ ) 4 to 20  $\mu\text{m}$  and contains a gibbsite type aluminium hydroxide according to claim 6 or a gibbsite type aluminium hydroxide obtained by a process according to any one of claims 1 to 5.



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8. A synthetic resin containing a gibbsite type aluminium hydroxide according to claim 6 or obtained by a process according to any one of claims 1 to 5.
9. The use of a gibbsite type aluminium hydroxide according to claim 6 or obtained by a process according to any one of claims 1 to 5 as a fire retardant additive in synthetic resins.

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(54) Title: PROCESS FOR THE PRODUCTION OF ALUMINIUM HYDROXIDE

(57) Abstract: Gibbsite type aluminium hydroxides are obtained by seeding a Bayer liquor with bayerite crystals. The gibbsite type aluminium hydroxides show excellent properties as fire retardant fillers in synthetic resins.

**RULE 63 (37 C.F.R. 1.63)**  
**DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION IN THE**  
**UNITED STATES PATENT AND TRADEMARK OFFICE**

As a below named inventor, I hereby declare that my residence, post office address and citizenship are as stated below next to my name, and I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the INVENTION ENTITLED "PROCESS FOR THE PRODUCTION OF ALUMINIUM HYDROXIDE" the specification of which was filed herewith as attorney docket No. 665-59079 and as US National Phase of PCT/EP00/05980.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose all information known to me to be material to patentability as defined in 37 C.F.R. 1.56. I hereby claim foreign priority benefits under 35 U.S.C. 119/365 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate filed by me or my assignee disclosing the subject matter claimed in this application and having a filing date (1) before that of the application on which priority is claimed, or (2) if no priority claimed, before the filing date of this application:

PRIOR FOREIGN APPLICATION(S)			Date first Laid- open or Published	Date Patented or Granted	Priority Claimed
Number	Country	Day/MONTH/Year Filed			Yes No
DE 99112401.7	Germany	29 June 1999			X

I hereby claim domestic priority benefit under 35 U.S.C. 120/365 of the indicated United States applications listed below and PCT international applications listed above or below and, if this is a continuation-in-part (CIP) application, insofar as the subject matter disclosed and claimed in this application is in addition to that disclosed in such prior applications, I acknowledge the duty to disclose all information known to me to be material to patentability as defined in 37 C.F.R. 1.56 which became available between the filing date of each such prior application and the national or PCT international filing date of this application:

PRIOR U.S. PROVISIONAL, NONPROVISIONAL AND/OR PCT APPLICATION(S)		Status	Priority Claimed
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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

I hereby appoint the registered practitioners represented by customer no.: 20736 of the law firm Manelli Denison & Selter, PLLC to prosecute this application and transact all business in the U.S. Patent and Trademark Office in connection therewith. Direct all correspondence to:

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